# Radiation efficiency of water-window Cherenkov sources using atomic-shell resonances

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A simple evaluation of the yield of Cherenkov radiation generated by relativistic electrons in the vicinity of atomic-shell resonances located in the water window x-ray domain is developed and applied to all the promising elements, including $L$-shell resonances explored recently by Knulst *et al.* [Appl. Phys. Lett. **83**, 4050 (2003)], and $K$-shell resonance in liquid nitrogen proposed here. Our theoretical results compare favorably with experimental data. The feasibility of a related Cherenkov laser is also studied. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850190]

The so-called water window (WW), a soft x-ray subdomain situated between the $K$-shell absorption edges of carbon ($E_C \approx 284.2$ eV, $\lambda_C = 43.62$ Å) and oxygen ($E_O \approx 543.1$ eV, $\lambda_O = 22.83$ Å), is of great importance to x-ray microscopy of live biological specimens. In WW, carbon in organic matter strongly absorbs the radiation, while water is almost transparent, thus providing strong contrast required for microscopy. So far, the primarily available source of radiation in WW has been synchrotron radiation, and alternative sources are of great interest.

It is known[1,2] that in a very narrow vicinity of their atomic-shell resonances (also called atomic absorption edges), a multilayer structure irradiated by relativistic electrons of moderate energy can generate strong transition radiation lines centered around absorption edges, due to pronounced peaks of the real part of refractive index for one of the constitutive components of the structure. In the soft x-ray domain these peaks can be large enough for the refractive index at the resonance to exceed 1, which in turn enables a single-layer material to radiate due to the Cherenkov effect within a narrow resonant line.

A very recent work by Knulst *et al.*[3] identified a group of elements (K, Ca, Sc, Ti, V) capable of exhibiting the Cherenkov effect at the $L_3$-shell atomic absorption edges within WW, and in a convincing elegant experiment demonstrated resonant radiation lines near $L_3$ shells for Ti and V foils irradiated by a 10-MeV electron beam. This source can be competitive with the existing ones. In view of its potential medical and bioapplications it is worth noting that the atomic-shell radiation lines from K, Ca, and N (see below) can serve also as a spectral tool for the imaging and measurements of the distribution of the respective elements in biospecimens; e.g., Ca is a major component of artery clogging, in particular in the human heart.

In this Letter, we (i) add to this list another element, nitrogen (to be used in a liquid phase), which is the only element in the periodic table situated between carbon and oxygen, and thus has its $K$ shell (instead of $L$ shells in the K, Ca, Sc, Ti, V) within WW, and (ii) propose a very simple way of evaluating the radiation efficiency, i.e., photon yield per electron, for all the candidate elements, which uses only three parameters for each element, to be easily found from published x-ray data (see, e.g., Refs. 4 and 5). Amazingly, this simple, almost back-of-the-envelope approach shows results that coincide very closely with the experimental data,[3] in fact even better than some of the theoretical evaluations in the same work. Our approach is valid also for any other x-ray radiation line based on the Cherenkov effect in the vicinity of atomic-shell resonances in any other domain where their refractive index $n$ exceeds 1.

When an electron passes through a medium with $n>1$, and its velocity $v=\beta c$ exceeds the phase velocity of light $c/n(\omega)$ at some frequency $\omega$, $\beta n(\omega)>1$, an electromagnetic (em) radiation at that frequency is emitted at the angle $\theta$ from $\vec{v}$ such that $\cos(\theta)=1/\beta n$. The photon yield, defined as the number of generated Cherenkov photons per electron, $N_{ph}$, can be evaluated in a transparent medium, based on Ref. 6, as

$$d^2N_{ph}/dkdz = \alpha f_C(k), \qquad (1)$$

where $k=\omega/c$ is the radiation wave number, $z$ is the propagation length, $f_C=\sin^2(\theta)=1-1/\beta^2 n^2>0$ is the Cherenkov factor, and $\alpha=e^2/\hbar c\approx 1/137$ is the fine-structure constant. In the optical domain, the Cherenkov effect is an efficient radiation tool enabling one, for example, to detect a single elementary relativistic particle. In the x-ray domain, however, the Cherenkov effect is rarely observable. Indeed, an absolutely necessary condition for the effect is that $\mathrm{Re}(n)>1$, while the prevailing situation in the x-ray domain is that $\mathrm{Re}(n)<1$. The physics behind this observation is the fact that for most of the x-ray spectra the dispersion of refractive index $\mathrm{Re}(n)$ is due to (effectively) unbound electrons and thus may be described by a plasmalike formula, $n^2\equiv\varepsilon=1-(\tilde{\omega}^2)_{K,L,\ldots}/\omega^2<1$. Here, $(\tilde{\omega}^2)_{K,L,\ldots}$, as in plasma, is proportional to the density of ("unbound" at the frequency $\omega$) electrons, whose number is related to the nearest (from below) atomic absorption edge, or shell resonance, $K,L,\ldots$. Fortunately, in the very close vicinities of those shell resonances, or the so-called atomic absorption edges, very dramatic changes of both the real and imaginary parts of $\varepsilon$ take place.

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When the photon energy becomes sufficient to remove electrons from a particular shell, the absorption jumps up almost discontinuously, hence the term "absorption edge." At the same time, since the newly "unbound" electrons abruptly assume a free-electron response to the radiation, the real part of $\varepsilon$ exhibits a sharp resonance with a narrow peak; the slopes of this peak are, however, not as steep, so that the dispersion line is broader than the absorption edge. Whereas absorption edges play a major role in various application of x rays, the resonances of the real part of $\varepsilon$ remains of little use to x-ray physics and applications, except for recently proposed resonant enhancement of the transition radiation in nanomultilayer solid-state structures[1,2] and Cherenkov radiation in WW.[3]

The fact most important for our purposes here is that, in the soft x-ray domain, the real part of refractive index $n$ (or the dielectric constant $\varepsilon=n^2$) in the very close vicinity of atomic-shell resonances may *exceed* 1, and that excess is sufficiently high (the magnitude of $\varepsilon-1$ for the above set of lines in WW can reach $\sim10^{-2}$) to allow for substantial Cherenkov radiation from electron beams of moderate energy.

Accounting for the x-ray absorption characterized by the attenuation length, $L_{at}(\omega)$, and integrating (1) over a finite thickness of a material layer, $d$, along $z$, we have

$$\frac{dN_{ph}}{dk} = \alpha f_C(k) L_{at}(k)\left\{1-\exp\left[-\frac{d}{L_{at}(k)}\right]\right\}. \tag{2}$$

Since in the x-ray domain we have $|n-1|\ll 1$ regardless of the sign of $n-1$, we need sufficiently high-energy electrons, $\gamma^2 \gg 1$, to excite Cherenkov radiation if $n-1>0$, where $\gamma = 1/\sqrt{1-\beta^2}$ is the relativistic factor. The formula for $f_C$ is reduced then to

$$f_C(k) \approx \Delta\varepsilon(k) - \gamma^{-2}, \quad \text{where}$$

$$\Delta\varepsilon = \mathrm{Re}(\varepsilon-1) \approx 2\,\mathrm{Re}(n-1). \tag{3}$$

The spectral lines of $\Delta\varepsilon(k)$ have unfamiliar shapes, involving terms like $\ln(k-k_{sh})$, $k_{sh}=cE_{sh}/\hbar$; $E_{sh}$ is the absorption edge energy and is different for different shells.[1,4,7,8] Detailed data for each element of the periodical table can now be found on various websites, see, e.g., Ref. 5. In the water window, the spectral linewidths of $\Delta\varepsilon(\omega)$ are about three orders of magnitude larger than the linewidths of the respective atomic absorption edges. Indeed, a relative atomic edge linewidth, $\Gamma_{at}=(2/3)k_{sh}r_e$, where $r_e=e^2/mc^2\sim 2.8\times10^5$ Å is the classical electron radius, so that at the WW midpoint, $\lambda\sim 33$ Å, we have $\Gamma\sim 0.35\times10^{-5}$, whereas typically, the relative linewidth, $\Delta k_{crs}/k_{sh}$ of the $\Delta\varepsilon(\omega)$ line at the crossover (see below) is $O(10^{-2})$. To estimate the maximum photon yield, we need to know, aside from the shell energy, $E_{sh}$, of each transition in question, three main characteristics for each of the six materials: the maximum magnitude of $\Delta\varepsilon_{mx}$ at that transition; the low crossover linewidth, $\Delta k_{crs}=k_{sh}-k_{low}$ (or $\Delta E_{crs}=\hbar c\Delta k_{crs}$), i.e., the spacing in $k$ space between the lower crossover point, $k_{low}$, at which $\Delta\varepsilon(k)=0$, and the absorption edge $k_{sh}$; and the averaged attenuation length, $\tilde{L}_{at}$, within that line below the point of the atomic absorption edge, i.e., below $E_{sh}$. If the electron relativistic factor $\gamma^2$ and the foil layer thickness, $d$, sufficiently exceed $1/\Delta\varepsilon_{mx}$ and $L_{at}$, respectively, the maximum photon yield (and the radiation spectrum) at each wave number does not depend on $\gamma$ and $d$ anymore, hence

$$d[N_{ph}(k)]_{mx}/dk = \alpha\Delta\varepsilon(k)L_{at}(k) \quad \text{for } \Delta\varepsilon(k)>0, \tag{4}$$

and the *total* maximum photon yield is $(N_\Sigma)_{mx} = \alpha\int_{k_{low}}^{k_{sh}}\Delta\varepsilon(k)L_{at}(k)dk$, or

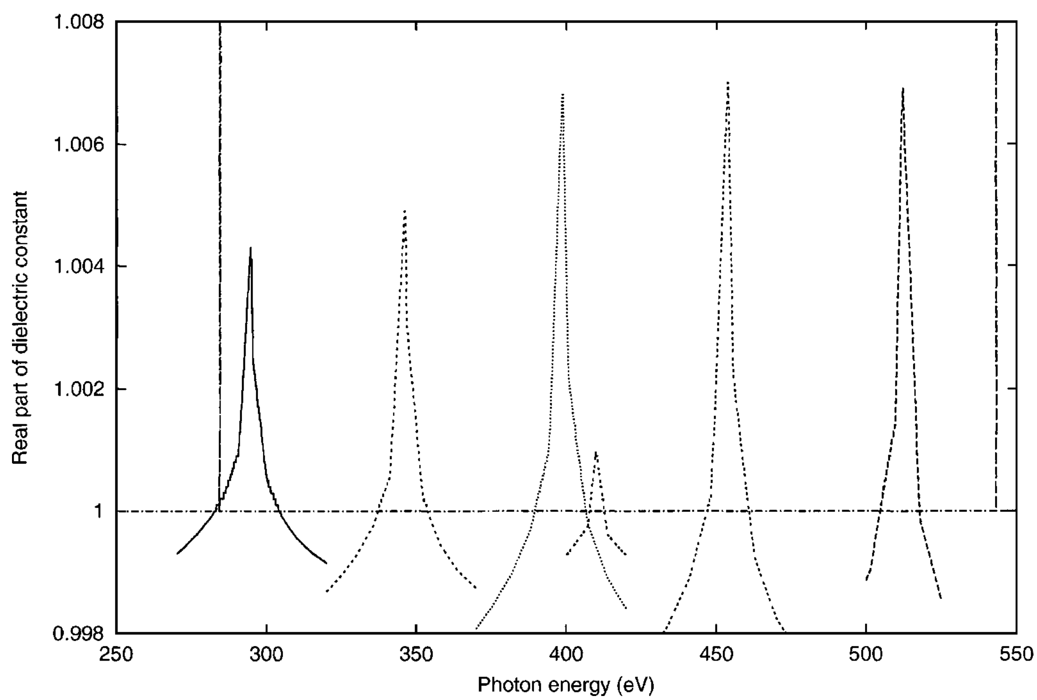


FIG. 1. Real part of dielectric constant $\varepsilon$ vs photon energy $E_{ph}$ eV for all the atomic-shell resonances in the water-window domain: K, Ca, Sc, N (liquid), Ti, and V, from left to right. Except for nitrogen with the $K$-shell line, all the rest of the materials have their resonances at $L_3$ shells. Vertical lines frame the water window: C on the left, O on the right.

TABLE I. Characteristics of atomic-shell resonances in the water window and their maximum total Cherenkov photon yields, $(N_\Sigma)_{mx}$.

| Element | K | Ca | Sc | N (liquid) | Ti | V |
|---|---|---|---|---|---|---|
| $\frac{E_{sh}}{\lambda_{sh}}\left(\frac{\text{eV}}{\text{Å}}\right)$ | $\frac{294.6}{42.1}$ | $\frac{346.2}{35.8}$ | $\frac{398.7}{31.1}$ | $\frac{409.9}{30.25}$ | $\frac{453.8}{27.3}$ | $\frac{512.1}{24.2}$ |
| $E_{crs}$ (eV) | 12 | 8.9 | 9.3 | 2.1 | 7 | 7.45 |
| $\Delta\varepsilon_{mx}\times 10^3$ | 4.3 | 4.91 | 6.8 | 0.95 | 7 | 6.87 |
| $L_{at}$ ($\mu$m) | 4.47 | 2.71 | 1.71 | 21.6 | 1.33 | 1.14 |
| $E_{thr}$ (MeV) | 7.28 | 6.78 | 5.69 | 16.07 | 5.6 | 5.65 |
| $(N_\Sigma)_{mx}\times 10^3$ | 2.59 | 2.19 | 2.0 | 0.797 | 1.206 | 1.08 |

$$(N_\Sigma)_{mx} \approx \alpha \Delta k_{crs} \Delta\varepsilon_{mx} \tilde{L}_{at}/2$$
$$\approx 1.85 \times 10^{-2} \Delta E_{crs}(\text{eV}) \Delta\varepsilon_{mx} \tilde{L}_{at}(\mu\text{m}). \quad (5)$$

In our estimation in (5) we neglected (i) all the photons with the energies above the absorption edge since their attenuation is substantially stronger [typically, in the edge vicinity, $L_{at}(k>k_{sh})$ is smaller than $\tilde{L}_{at}$ by an order of magnitude], and (ii) the fact that the dispersion of $\varepsilon(k)$ between the absorption edge and the lower crossover point is not quite linear (see Fig. 1). Fortunately, these two effects are not significant, and furthermore, have their contributions with opposite sign, so to a great degree they cancel each other. The spectra of $\Delta\varepsilon(E)$ in terms of photon energy, $E=\hbar\omega$, for all six materials are shown in Fig. 1. Note that the $L_3$-shell lines of all elements under consideration except for nitrogen are noticeably broadened, due to a contribution of the nearby (higher) $L_2$ shell. This is, however, insignificant for our purposes, because the $L_2$-absorption edge is almost nonexistent, and because the fact that $\Delta\varepsilon>0$ above the $L_3$ transition is almost lost for our purposes, since in that area, the radiation is strongly inhibited by the attenuation from $L_3$ edge anyway. All the relevant parameters and characteristics are found in Table I. Note that, although nitrogen has a noticeably lower magnitude of $\Delta\varepsilon_{mx}$ at its $K$ transition than other materials do, its photon yield, $(N_\Sigma)_{mx}$, is of the same order of magnitude as the yield for the rest of the candidates; this is due to a much longer attenuation length, $L_{at}$, of N.

$(N_\Sigma)_{mx}$ is the *maximum* yield attained at $\gamma^2 \gg 1/\Delta\varepsilon_{mx}$; less energetic $e$ beams engage only part of the Cherenkov line. An estimate for the yield $N_\Sigma$ for any $\gamma$ is as

$$N_\Sigma/(N_\Sigma)_{mx} \approx (1-\gamma_{thr}^2/\gamma^2)^2 = [1-1/(\gamma^2\Delta\varepsilon_{mx})]^2, \quad (6)$$

where $\gamma_{thr}=1/\sqrt{\Delta\varepsilon_{mx}}$ is the threshold relativistic factor to excite Cherenkov radiation; for the respective energies of $e$ beam, $E_{thr}=mc^2(\gamma_{thr}-1)$ see Table I. Thus our yield estimates for Ti and V at $E=10$ MeV are 5.75 and 4.645 $\times 10^{-4}$, respectively, which comes much closer to the respective experimental data for V ($4.3\times 10^{-4}$) than the theoretical evaluation[3] of $1.4\times 10^{-4}$, and within the same range from experimental data[3] for Ti ($3.5\times 10^{-4}$) as the theoretical evaluation[3] of $2.4\times 10^{-4}$.

It is tempting to look into the feasibility of a WW Cherenkov laser based on atomic-shell resonances. Our preliminary investigation shows that electron density in the regular accelerators, $\rho$, required for the gain, $G$, to exceed the losses in the system (due to attenuation of Cherenkov radiation in a sample and absorption and transmission in mirrors, in particular) is hardly available at the moment. Using the stimulated Cherenkov gain, $G$,[9] simplified for $\gamma \gg 1$ and $n\sim 1$ as $G\approx (r_e\lambda^2)\Delta\varepsilon\gamma^{-1}\rho A$, where

$$A = 2\pi^2(L/\lambda)^3\theta^2(d/du)[\sin(u)/u]^2,$$
$$u = (\pi L/2\lambda)(\gamma^{-2}+\theta^2-\Delta\varepsilon) \quad (7)$$

we estimate a minimal electron density $\rho_{\min}$ to attain the gain $G$ at the length $L$ as

$$\rho_{\min} = \text{O}(1) \times G \times (\lambda/r_e L^3)\gamma/\Delta\varepsilon^2. \quad (8)$$

For the data in Table I, Eq. (8) yields an estimate for $\rho$ needed for $G>1$ at $L\sim\tilde{L}_{at}$, as $\rho\sim 4\times 10^{21}$ cm$^{-3}$. Relativistic electron currents with such density could conceivably be attained only in the focal area of relativistically intense lasers in solid state.

In conclusion, we developed a simple way of evaluating the radiation efficiency of Cherenkov radiation by relativistic electrons in the water-window x-ray domain in the sources based on atomic-shell resonances for all the materials having significant WW resonances (including liquid nitrogen proposed here); the results show favorable comparison with available experimental data.

[1] A. E. Kaplan, C. T. Law, and P. L. Shkolnikov, Phys. Rev. E **52**, 6795 (1995).
[2] K. Yajima, T. Awata, R. Koizumi, M. Imai, A. Itoh, and N. Imanishi, Nucl. Instrum. Methods Phys. Res. A **435**, 490 (1999).
[3] W. Knulst, M. J. van der Viel, O. J. Luiten, and J. Verhoeven, Appl. Phys. Lett. **83**, 4050 (2003).
[4] B. L. Henke, E. M. Gullikson, and J. C. Davis, At. Data Nucl. Data Tables **54**, 181 (1993).
[5] A greatly useful site "X-Ray Interactions With Matter" run by E. M. Gullikson at http://www-cxro.lbl.gov/optical_constants/ provides tables and on-line calculations in the 50 eV–30 KeV x-ray domain based mostly on Ref. 4, it also has links to other x-ray-related Internet sources.
[6] Ig. Tamm, J. Phys. (Moscow) **1**, 439 (1939); J. V. Jelley, *Cherenkov Radiation and its Applications* (Pergamon, London, 1958); M. L. Ter-Mikaelian, *High Energy Electromagnetic Processes in Condensed Media* (Wiley Interscience, New York, 1972).
[7] L. G. Parratt and C. F. Hampstead, Phys. Rev. **94**, 1593 (1954).
[8] The physics of resonant anomalous x-ray dispersion and scattering has been developed in great detail in the context of crystallography. A recent collection of articles can be found in "*Resonant Anomalous X-ray Scattering. Theory and Applications*," edited by G. Materlik, C. J. Sparks, and K. Fisher (North-Holland, Amsterdam, 1994), in particular, the reviews by B. Lengeler, p. 35, R. L. Blake, J. C. Davis, D. E. Graessle, T. H. Burbine, and E. M. Gullikson, p. 79, and D. H. Templeton, p. 1.
[9] W. Becker and J. K. McIver, Phys. Rev. A **31**, 783 (1985). A. E. Kaplan and S. Datta, Appl. Phys. Lett. **44**, 661 (1984), S. Datta and A. E. Kaplan, Phys. Rev. A **31**, 790 (1985).